PATENT SPECIFICATION

1,118,597

NO DRAWINGS

1,118,597

Date of Application and filing Complete Specification: 4 Nov., 1965.
No. 46723/65.
Application made in Germany (No. C34293 IVc/22i) on 5 Nov., 1964.
Application made in Germany (No. C34643IVc/22i) on 15 Dec., 1964.
Complete Specification Published: 3 July, 1968.

Index at acceptance:—B5 K3

Int. Cl.:— C 09 j 5/00

COMPLETE SPECIFICATION

Process for Uniting Components of Polybutene-(1)

We, CHEMISCHE WERKE HÜLS AKTIENGESELLSCHAFT, a German Company, of 4370 Marl, Germany, do hereby declare the invention, for which we pray that a Patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

It is known that components of polyolefines cannot be joined together by adhesion or can only be joined together very imperfectly. For this reason it is necessary to use other jointing methods, for example heat sealing; screw joints and flanged joints may be used for pipes.

Since adhesion requires the least expenditure, however, attempts have repeatedly been made to solve this problem. Hagen and Domininghaus, in "polyäthylen und andere Polyolefine", 2nd edition, 1961, published by B. Garrels, Hamburg, summarise in page 348 that non-polar polyolefines behave unfavourably in adhesion so that the usual way in the case of many plastics of treating the surfaces with a solvent and uniting them under pressure is unsuccessful in the case of polyolefines. Polyolefines include in this connection not only polyethylene and polypropylene but also polybutene-(1).

It is known that polyisobutylene may be stuck (latest W. Hawerkamp, Chem, Ind. *16* (1964) No. 6, 329) but at the same place the usual statements concerning the poor adhesion of polyethylene and polypropylene are to be found so that the existing prejudice regarding poly-n-butene is not invalidated. It is known that nothing can be concluded as to polybutene-(1) from the properties of polyisobutylene; the latter, as a polymer of an isoolefine, is elastomeric, the former, as a member of n-olefines polymerised in the chain, is a rigid plastic.

Thus it is surprising in the highest degree that it should be possible to adhere polybutene-(1) very well.

We have found that components of polybutene-(1) can be united by treating the components with a liquid hydrocarbon, which may be substituted or polymerisable and which may contain polybutene-(1) dissolved therein, and pressing the components together at temperatures of 50°C to 100°C.

Polybutene-(1) is a plastic which is obtained from butene-(1) by one of the conventional methods of polymerisation, particularly by the low pressure polymerisation method of Ziegler.

Components capable of being adhered are for example both semi-finished products, such as pipes, sections, sheets, plates and films and also finished products of all types which are brought into their final form or are repaired by an adhesion method.

Examples of suitable liquid hydrocarbons are paraffins, such as heptane, gasoline, diesel oil or paraffin oil; aromatics, such as benzene or xylene; cycloaliphatics such as cyclohexane, isopropylcyclohexane; alkylaromatics, such as tetrapropylenebenzene or dodecylbenzene; and also substituted hydrocarbons, such as carbon tetrachloride or chlorobenzene. Alkylbenzenes are preferred, particularly tetrapropylenebenzene or dodecylbenzene.

Examples of suitable liquid polymerisable hydrocarbons are styrene or α-methylstyrene. These substances, in addition to adhesion, effect a permanent joint in a most advantageous way by autopolymerisation.

To achieve a more rapid polymerisation, 0.001 to 1%, preferably 0.01 to 0.2%, of a radical-forming compound may be added to the polymerisable hydrocarbon, for example peroxides, such as benzoyl peroxide, lauroyl peroxide, capryl peroxide, isopropyl percarbonate, p-menthane hydroperoxide or azo compounds, such as azoisobutyrodinitrile or azodivalerodinitrile.

For the convenient lowering of the treatment temperature, is is recommendable to add sensitisers, such as sulphites, thiosulphates, hydrazine, mercaptan, methanol, oxalic acid,

polyhydric alcohols or ferrous sulphate, to the radical-forming compounds if necessary. These sensitisers are used in amounts of 0.1 to 1000%, preferably 0.5 to 250%, with reference to the radical-forming compound.

These liquid hydrocarbons may contain polybutene-(1) dissolved therein, advantageously in concentrations of about 1 to 10% by weight; for example an about 5% solution of polybutene-(1) in heptane or benzene is very useful.

The liquid hydrocarbon may be applied in a conventional way by brushing on, by dipping the components in the hydrocarbon or by other suitable means.

In some cases, particularly when using a solvent containing polybutene-(1) as adhesive, it is sufficient to pretreat only one of the two parts to be united with the adhesive solution. In general however quite particularly durable joints can be achieved by pretreating both components. Before the components are brought together, especially when using a high boiling point solvent, they may be kept in the air for about one to ten minutes so that a preliminary swelling of the adhesive surfaces and a partial evaporation of the excess solvent takes place.

During bonding, the components to be joined are advantageously exposed to a pressure which may be from 5 to 20, preferably 5 to 10, atmospheres gauge. Higher or lower pressures are naturally also possible, but as a rule do not bring any advantages, provided no special reasons exist, as for example too thin wall thickness of the components to be joined.

As a rule it is sufficient to maintain a pressure of 10 atmospheres gauge for ten minutes at a temperature of 50°C when bonding relatively thin-walled components, such as pipes, using dodecylbenzene as adhesive; the time required is shorter at higher temperature and also when using a solvent having a lower boiling point.

A preferred embodiment of the process consists in treating the surfaces to be united with a heated, for example boiling, solvent. The desired pressure may then be applied in known manner for example by driving an end of a pipe which has been pretreated externally with solvent into a somewhat widened end which has been pretreated internally.

Bonds whose tensile strength is not less than that of the bonded material may be directly prepared by means of the new adhesion method. Particularly good results are achieved when mouldings of polybutene-(1) are bonded soon after they have been made and while the plastic is still in the unstable modification II.

It may be considered as particularly surprising that this result is achieved, contrary to all expectation, even by simple partial solution of the surface with a suitable solvent.

The adhesion experiments given in the Examples are carried out as press-moulded plates having a thickness of 1 mm. The strengths of the press-moulded plates and of the adhesive joints are measured in each case after ageing for ten days at room temperature so that comparable values of the stable modification I of the polybutene are shown. In the Tables, E = Example; P = pressure: T = temperature; ATSJ = absolute tensile strength (kg/1.5 cm specimen width) at the joint; ATSJ/P = ratio of absolute tensile strength at the joint to the absolute tensile strength of the press-moulded plate.

EXAMPLE 1 TO 5

Two freshly prepared press-moulded plates of polybutene having an absolute tensile strength of 15.0 kg/1.5 cm specimen width and a thickness of 1 mm are coated with a 5% solution of polybutene in heptane, overlapped by 1 cm and then pressed for ten minutes at the pressures and temperatures given in the Table:

E	P	T	ATSJ	ATSJ/P
1	10	60° C.	10.4	0.69
2	10	70° C.	10.5	0.70
3	10	80° C.	10.8	0.72
4	10	90° C.	12.9	0.86
5	5	100° C.	9.4	0.63

EXAMPLE 6 TO 11

Two press-moulded plates of polybutene (aged for ten days at room temperature) and having an absolute tensile strength of 15.0 kg/1.5 cm of specimen width and a thickness of 1 mm are thinly coated with the solvents specified in the Table, overlapped by 1 cm and then pressed together for ten minutes at 20 atmospheres gauge and 100°C.

E	Solvent	ATSJ	ATSJ/P
6	benzene	12.4	0.83
7	tetrapropylenebenzene	14.3	0.95
8	alkylbenzene (alkyl = C_{12})	15.7	about 1.00
9	paraffin oil	13.0	0.87
10	chlorobenzene	12.4	0.83
11	carbon tetrachloride	13.9	0.93

EXAMPLE 12 TO 17

The next Examples correspond to Examples 6 to 11 but are carried out on freshly prepared press-moulded plates. For these experiments a polybutene having a higher absolute tensile strength of 23.5 kg/1.5 cm of specimen width is used.

E	Solvent	ATSJ	ATSJ/P
12	benzene	23.7	about 1.0
13	tetrapropylenebenzene	22.4	0.95
14	alkylbenzene (alkyl = C_{12})	21.1	0.9
15	paraffin oil	22.1	0.94
16	chlorobenzene	23.5	0.99
17	carbon tetrachloride	23.2	0.99

EXAMPLES 18 TO 20

Two press-moulded plates of polybutene having an absolute tensile strength of 23.5 kg/1.5 cm specimen width, which have been aged for ten days at room temperature, are dipped for a short time in paraffin oil which has been heated to 120°C, overlapped by 1 cm and then pressed together for ten minutes at 5 atmospheres gauge. TP = temperature of paraffin oil; TB = temperature during bonding.

E	TP	TB	ATSJ	ATSJ/P
18	120° C.	100° C.	24.0	about 1.00
19	120° C.	90° C.	20.6	0.88
20	130° C.	100° C.	25.3	about 1.00

EXAMPLE 21 TO 23

Two press-moulded plates of polybutene-(1) having an absolute tensile strength of 26.3 kg/1.5 cm specimen width and a thickness of 1 mm are coated with styrene at 90°C, overlapped by 1 cm and then pressed for ten minutes at the temperatures and pressures given in the Table. The joint obtained is not swollen and is therefore true to size.

E	P	T	ATSJ	ATSJ/P
21	10	90° C.	20.1	0.76
22	10	100° C.	23.9	0.91
23	20	100° C.	25.3	0.96

EXAMPLE 24 TO 26

Two press-moulded plates of polybutene-(1) having an absolute tensile strength of 26.3 kg/1.5 cm specimen width and a thickness of 1 mm are coated with styrene and the activators specified in the Table, overlapped by 1 cm and then pressed together for ten minutes at 10 atmospheres gauge and 90°C. Components having hard joints and accurate trueness to size.

E	Activator	ATSJ	ATSJ/P
24	0.1% benzoyl peroxide	24.5	0.93
25	0.1% azodiisobutyrodinitrile	24.2	0.92
26	0.1% lauroyl peroxide and 0.2% methanol	25.0	0.95

EXAMPLE 27

Two press-moulded plates of polybutene-(1) having an absolute tensile strength of 26.3 kg/1.5 cm specimen width and a thickness of 1 mm are coated with α-methylstyrene which contains 0.1% of capryl peroxide, overlapped by 1 cm and then pressed together for ten minutes at 10 atmospheres gauge and 90°C. The absolute tensile strength of the joint is 23.7 kg/1.5 cm specimen width. The ratio of the absolute tensile strength of the joint to the absolute tensile strength of the press-moulded plates is 0.90.

WHAT WE CLAIM IS:—

1. A process for bonding components of polybutene-(1) wherein the components are treated with a liquid hydrocarbon and then joined together under pressure at a temperature of 50° to 100°C.

2. A process as claimed in claim 1 wherein the liquid hydrocarbon is a substituted hydrocarbon.

3. A process as claimed in claim 1 or 2 wherein the liquid hydrocarbon is a polymerisable hydrocarbon.

4. A process as claimed in any one of claims 1 to 3 wherein the liquid hydrocarbon contains polybutene-(1) dissolved therein.

5. A process as claimed in claim 3 or 4 wherein the hydrocarbon contains a radical-forming compound in an amount of 0.001 to 1% by weight.

6. A process as claimed in claim 5 wherein the said percentage is 0.01 to 0.2%.

7. A process as claimed in any one of claims 3 to 6 wherein the hydrocarbon contains a sensitiser in an amount of 0.1 to 1000% on the radical-forming compound.

8. A process as claimed in claim 7 wherein the said percentage is 0.5 to 250%.

9. A process as claimed in any one of claims 1 to 8 wherein the pressure used is from 5 to 20 atmospheres gauge.

10. A process as claimed in claim 9 wherein the said pressure is 5 to 10 atmospheres gauge.

11. A process as claimed in claim 1 carried out substantially as described in any of the foregoing Examples.

12. Bonded components of polybutene-(1) when obtained by the process claimed in any of claims 1 to 11.

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Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1968.
Published by the Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.